Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

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To cite this Article Haraya, Kenji , Hakuta, Toshikatsu and Yoshitome, Hiroshi(1985) 'The Calculation of a Serial Fed Gas Permeator System', Separation Science and Technology, 20: 5, 403 — 421
To link to this Article: DOI: 10.1080/01496398508060690
URL: http://dx.doi.org/10.1080/01496398508060690

SEPARATION SCIENCE AND TECHNOLOGY, **20**(5 & 6), pp. 403-421, 1985

The Calculation of a Serial Fed Gas Permeator System

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Abstract

The calculation methods for the serial fed multistage system were developed for the four flow pattern, i.e., perfect mixing, cocurrent flow, cross flow, and countercurrent flow, under the condition of constant membrane area or constant cut. The calculations were performed for two examples using permeabilities of polyimide. One is the case in which concentrate H_2 is up to 95% in the total permeate stream from a 75% H_2–25% CO mixture. Another is the case in which concentrate CH_4 is up to 98% in the high-pressure stream from a 60% CH_4–40% CO_2 mixture. A brief parametric study shows that the system consisting of countercurrent flow modules under the condition of constant cut is most efficient among the case studied and should minimize the number of stages using big modules. On the other hand, the system composed of perfect mixing or cocurrent flow modules under the condition of constant membrane area is more efficient than constant cut, and it is advantageous to assemble a system using a large number of small modules. The performance of the system comprised of cross flow modules is identical with that of a single module.

INTRODUCTION

With the advance of membrane fabrication technology, the use of membrane is becoming economically attractive for gas separation processes. Recently, many membrane separators have been used commercially for recovery of hydrogen in ammonia, petroleum refining, and petrochemical plants, as well as for separation of carbon dioxide from biogas and hydrocarbons for enhanced oil recovery (1–5).

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Multiple module element systems are adopted in these industrial applications when the area requirement exceeds that of a single module (*6*). Common configurations include parallel and serial fed module arrangements. In the parallel arrangement, slight differences in resistance to flow in any parallel path causes distribution of the flow rate among the modules. Poor distribution in a parallel system results in declines in both product purity and recovery. That system must be designed and operated to have uniform distribution by flow rate regulators. The serial fed arrangement does not require particular attention to flow rate distribution. It is advantageous to select the serial fed system for design and operation. In this arrangement, however, the feed stream composition of each module varies from top to tail. The total performance of this system is predicted by stepwise calculation from beginning to end.

McCandless (*7*) made calculations on a system consisting of perfect mixing modules and Hwang and Kammermeyer (*8*) also made calculations on a system composed of perfect mixing or cocurrent flow modules. Those reports didn't contain sufficient information to understand the serial fed system. Specifically, the relationships of the operating parameters between each module forming the system were not characterized.

Once the desired value (i.e., product purity or recovery, etc.) is determined, the prediction of the serial fed arrangement is a complicated iterative calculation, because total performance is dependent on the membrane area and the stage cut of each module. It is necessary to formulate the operating relationship between modules for iterative calculation.

In the following, calculation methods are presented for the separation of a binary gas by using a serial fed arrangement system which consists of four kinds of ideal flow patterns, i.e., perfect mixing, cocurrent, cross, and countercurrent flow. The methods are useful for the design and prediction of performance of the system.

THEORY

Total Material Balance

Figure 1 illustrates a serial fed arrangement system in which high-pressure streams are connected in series and permeate streams are collected through parallel paths.

The cut of the *n*th stage is defined as

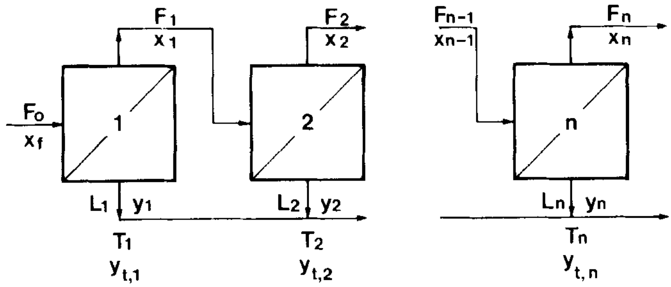

FIG. 1. Diagram of serial fed multiple stage system.

$$\Theta = L_n/F_{n-1} \tag{1}$$

The flow rates of high-pressure and permeate streams are as follows.

Inlet of high-pressure stream:

$$\begin{aligned} F_{n-1} &= F_0(1-\Theta_1)(1-\Theta_2)\cdots(1-\Theta_{n-1}) \\ &= F_0\prod_{i=1}^{n-1}(1-\Theta_i) \end{aligned} \tag{2}$$

Outlet of high-pressure stream:

$$F_n = F_0\prod_{i=1}^{n}(1-\Theta_i) \tag{3}$$

Permeate stream:

$$L_n = F_0\Theta_n\prod_{i=1}^{n-1}(1-\Theta_i) \tag{4}$$

Total permeate stream:

$$T_n = \sum_{j=1}^{n}\left\{F_0\Theta_j\prod_{i=1}^{j-1}(1-\Theta_i)\right\} \tag{5}$$

The total material balance around the first and nth stages is

$$F_0 = F_0\prod_{i=1}^{n}(1-\Theta_i) + \sum_{j=1}^{n}\{F_0\Theta_j\prod_{i=1}^{j-1}(1-\Theta_i)\} \tag{6}$$

$$x_fF_0 = x_nF_0\prod_{i=1}^{n}(1-\Theta_i) + y_{t,n}\sum_{j=1}^{n}\left\{F_0\Theta_j\prod_{i=1}^{j-1}(1-\Theta_i)\right. \tag{7}$$

Transformation of Eq. (7) yields

$$y_{t,n} = \frac{x_f F_0 - x_n F_0 \prod_{i=1}^{n} (1 - \Theta_i)}{\sum_{j=1}^{n} \left\{ F_0 \Theta_j \prod_{i=1}^{j-1} (1 - \Theta_i) \right\}} \tag{8}$$

Total cut Θ_t is

$$\Theta_t = T_n/F_0 = \sum_{j=1}^{n} \left\{ \Theta_j \prod_{i=1}^{j-1} (1 - \Theta_i) \right\} \tag{9}$$

When the stage cut is constant in any stage ($\Theta_n = \Theta$), F_n and T_n are simplified to

$$F_n = F_0(1 - \Theta)^n \tag{10}$$

and

$$T_n = \Theta \sum_{i=1}^{n-1} F_0(1 - \Theta)^i = F_0\{1 - (1 - \Theta)^n\} \tag{11}$$

Accordingly, Eqs. (8) and (9) become

$$y_{t,n} = \frac{x_f - x_n(1 - \Theta)^n}{1 - (1 - \Theta)^n} \tag{12}$$

and

$$\Theta_t = 1 - (1 - \Theta)^n \tag{13}$$

Mathematical Formulation of Single Stage

The composition of the reject stream x_n in Eq. (8) or Eq. (12) is dependent on the feed and permeate flow pattern (perfect mixing, cocurrent, cross flow, or countercurrent), and various calculation methods have been reported in the literature (*7–14*). The derivations of the model equations in this paper are essentially the same as those reported previously except for the expression of the membrane area.

The following mathematical formulation for the permeation of a binary gas in terms of various flow patterns is based on three assumptions:

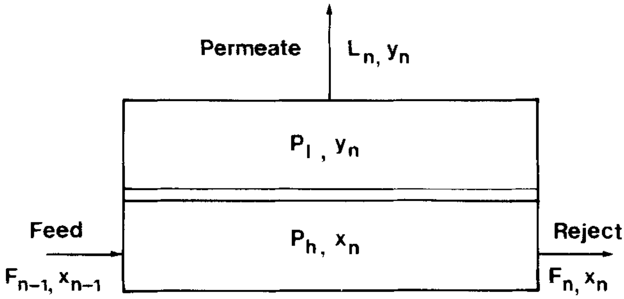

FIG. 2. Single stage with perfect mixing.

(1) Membrane permeabilities are independent of pressure and concentration.

(2) Pressure drops are negligible in the high-pressure stream and in the permeate stream in their bulk flow directions.

(3) Diffusion in the permeate direction and in the bulk flow direction is negligible.

Perfect Mixing

First, we consider the perfect mixing mode where the concentration and pressure are almost uniform in both high-pressure and permeate streams as illustrated in Fig. 2. The permeation equations are derived as

$$L_n y_n = \frac{Q_A p_h}{l} S_n(x_n - Pr_n y_n) \tag{14}$$

$$L_n(1 - y_n) = \frac{Q_B p_h}{l} S_n\{1 - x_n - Pr_n(1 - y_n)\} \tag{15}$$

where Pr_n is the ratio of low (permeate) to high pressure (p_l/p_h). Combining Eqs. (14) and (15), one obtains

$$\frac{y_n}{1 - y_n} = \alpha^* \frac{x_n - Pr_n y_n}{1 - x_n - Pr_n(1 - y_n)} \tag{16}$$

From the material balance,

$$x_n = \frac{x_{n-1} - \Theta_n y_n}{1 - \Theta_n} \tag{17}$$

Solving for x_n from Eqs. (16) and (17) yields

$$x_n = \frac{x_{n-1}}{1 - \Theta_n} - \frac{\Theta_n\{B - (B^2 - AC)^{1/2}\}}{A(1 - \Theta_n)} \tag{18}$$

where

$$A = (\alpha^* - 1)\{\Theta_n + Pr_n(1 - \Theta_n)\}$$
$$B = 0.5[(\alpha^* - 1)\{\Theta_n + Pr_n(1 - \Theta_n) + x_{n-1}\} + 1]$$
$$C = \alpha^* x_{n-1}$$

The membrane area becomes

$$S_n = \frac{lF_{n-1}}{Q_A p_h} \frac{\Theta_n y_n}{x_n - Pr_n y_n}$$
$$= \frac{lF_0}{Q_A p_h} \frac{\Theta_n y_n}{x_n - Pr_n y_n} \prod_{i=1}^{n-1} (1 - \Theta_n) \tag{19}$$

Introducing the dimensionless membrane area (*8*) St_n as

$$St_n = S_n/S^* \tag{20}$$

where S^* is defined as

$$S^* = \frac{F_0 l}{Q_A p_h} \tag{21}$$

then St_n becomes, from Eq. (19),

$$St_n = \frac{\Theta_n y_n}{x_n - Pr_n y_n} \prod_{i=1}^{n-1} (1 - \Theta_i) \tag{22}$$

When the cut is constant in all stages, St_n is simplified to

$$St_n = \frac{\Theta y_n}{x_n - Pr_n y_n} (1 - \Theta)^{n-1} \tag{23}$$

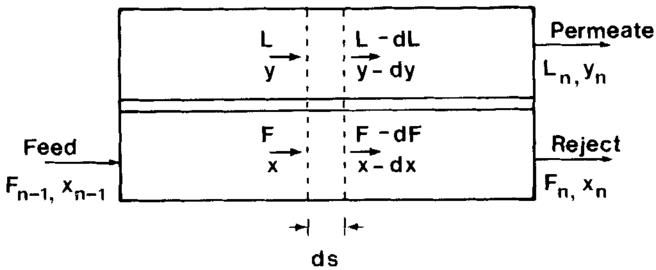

FIG. 3. Single stage with cocurrent flow.

The total membrane area from the 1st to the nth stage is the product of S^* and ΣSt_n.

Cocurrent, Countercurrent, and Cross Flow

Figure 3 illustrates a cocurrent flow pattern. In the case of cocurrent flow, assuming that plug flow exists in the high-pressure and permeate streams, the permeation equations are derived over a differential membrane area dS as

$$-d(Fx) = \frac{Q_A p_h}{l} dS(x - Pr_n y) \tag{24}$$

and

$$-d[F(1 - x)] = \frac{Q_B p_h}{l} dS[1 - x - Pr_n(1 - y)] \tag{25}$$

Equations (24) and (25) can be transformed to

$$\frac{dx}{dF'} = \frac{(x - Pr_n y) - x[x - Pr_n y + (1/\alpha^*)\{(1 - x) - Pr_n(1 - y)\}]}{F'[x - Pr_n y + (1/\alpha^*)\{(1 - x) - Pr_n(1 - y)\}]} \tag{26}$$

and

$$\frac{dS_d}{dF'} = \frac{-1}{x - Pr_n y + (1/\alpha^*)\{1 - x - Pr_n(1 - y)\}} \tag{27}$$

where the following dimensionless variables are introduced:

$$F' = \frac{F}{F_{n-1}} \tag{28}$$

$$S_d = \frac{Q_A p_h}{lF_{n-1}} S \tag{29}$$

Using F_0, S_d is expressed as

$$S_d = \frac{S}{S^* \prod_{i=1}^{n-1} (1 - \Theta_i)} \tag{30}$$

From the material balance, y is given by

$$y = \frac{x_{n-1} - F'x}{1 - F'} \qquad (F' \neq 1) \tag{31}$$

The system of differential equations is numerically integrated with the initial conditions

$$x = x_{n-1},\ y = y_i,\ S_d = 0 \text{ at } F' = 1$$

so that

$$x = x_n,\ y = y_n,\ S_d = S_{d,n} \text{ at } F' = (1 - \Theta_n)$$

y_i is determined from

$$\frac{y_i}{1 - y_i} = \frac{\alpha^*(x_{n-1} - Pr_n y_i)}{1 - x_{n-1} - Pr_n(1 - y_i)} \tag{32}$$

It is convenient to use the modified dimensionless membrane area St_n:

$$St_n = S_{d,n} \prod_{i=1}^{n-1} (1 - \Theta_i) \tag{33}$$

When the cut is constant in all stages,

$$St_n = S_{d,n}(1 - \Theta)^{n-1} \tag{34}$$

the actual membrane area of N stages is the product of S^* and ΣSt_n.

In countercurrent flow, Eqs. (26) and (27) are still valid, but Eq. (31) is replaced by

$$y = \frac{F'x - x_n(1 - \Theta_n)}{F' - (1 - \Theta_n)} \qquad (F' \neq 1 - \Theta_n) \tag{35}$$

Accordingly, Eqs. (26), (27), and (35) are numerically integrated backward with the initial conditions

$$x = x_n,\ y = y_0,\ S_d = S_{d,n} \text{ at } F' = (1 - \Theta_n)$$

to find

$$x = x_{n-1},\ y = y_n,\ S_d = 0 \text{ at } F' = 1$$

where y_0 is determined from Eq. (32) by substituting x_n for x_{n-1} and y_0 for y_i. This calculation requires that an initial value x_n be assumed and that an iteration procedure be applied until x_{n-1} has the same value as the inlet composition.

In the cross flow, the permeate flow is mainly in the direction normal to the membrane. Equations (26) and (27) are still valid, and

$$\frac{d(Fx)}{dF} = y \tag{36}$$

is added. With the aid of Eq. (36), the ratio of Eqs. (24) and (25) becomes

$$\frac{y}{1 - y} = \frac{\alpha^*(x - Pr_n y)}{1 - x - Pr_n(1 - y)} \tag{37}$$

Accordingly, Eqs. (26), (27) and (37) are integrated with the initial conditions

$$x = x_{n-1},\ S_d = 0 \text{ at } F' = 1$$

to find

$$x = x_n,\ y = y_n,\ S_d = S_{d,n} \text{ at } F' = (1 - \Theta_n)$$

TABLE 1
Permeabilities of Various Gases through the Polyimide (25°C)

Gas	Permeability [$m^3(STP) \cdot m \cdot m^{-2} \cdot s^{-1} \cdot Pa^{-1}$]	Permeability ratio α^*	
H_2	3.90×10^{-17}		
CO	5.12×10^{-19}	H_2/CO	76.2
CO_2	1.11×10^{-17}		
CH_4	2.16×10^{-19}	CO_2/CH_4	51.4
O_2	1.90×10^{-18}		
N_2	2.44×10^{-19}	O_2/N_2	7.75

Calculation Procedure

Using the equations described above, the total performance of the serial fed system consisting of N stages can be predicted for given x_f, α^*, Pr_n, and Θ_n by repeatedly calculating the single stage from the first to the Nth. However, designing an N stages system which attains the desired product purity, product recovery, or total cut generally requires a trial and error procedure. In the following, calculation procedures are described for several cases in which it is assumed that Pr_n is constant in all stages. The special cases selected here have a constant cut and a constant membrane area, and are of practical interest.

Case 1. Constant Membrane Area

(1) As a first step, assume Θ_1 and calculate x_1 by Eq. (17) or integrate Eqs. (26) and (27) numerically. Then calculate St_1 by Eq. (22) or Eq. (33).

(2) Assume Θ_2 and calculate x_2 and St_2 by a procedure similar to Step (1). Repeat this procedure until St_2 becomes close to St_1 within the desired accuracy.

(3) Repeat a calculation similar to Step (2) from the 2nd to the Nth stages and then calculate x_N, $y_{t,N}$ (by Eq. 8), product recovery, or Θ_t (by Eq. 9).

(4) Repeat Steps (1) to (3) until x_N, $y_{t,N}$, product recovery, or Θ_t becomes close to the desired value within the desired accuracy.

TABLE 2
Hydrogen Separation from a H_2/CO Gas Mixture Using a Serial Fed Separator ($\alpha^* = 76.2$, $x_f = 0.75$, $N = 4$, constant membrane area)

Mode	n	x_n	y_n	Θ_n	St_n	Pr
Perfect mixing[a]	1	0.6050	0.9838	0.3814	1.208	0.3
	2	0.4425	0.9561	0.3180	1.208	0.3
	3	0.3285	0.8836	0.2054	1.208	0.3
	4	0.2592	0.7673	0.1364	1.208	0.3
Cocurrent[b]	1	0.5503	0.9880	0.4563	1.241	0.3
	2	0.3410	0.9513	0.3429	1.241	0.3
	3	0.2576	0.8022	0.1532	1.241	0.3
	4	0.2090	0.6594	0.1079	1.241	0.3
Cross flow[c]	1	0.5464	0.9880	0.4611	1.255	0.3
	2	0.3274	0.9515	0.3508	1.255	0.3
	3	0.2348	0.8069	0.1619	1.255	0.3
	4	0.1853	0.6440	0.1079	1.255	0.3
Countercurrent[d]	1	0.5434	0.9879	0.4649	1.266	0.3
	2	0.3183	0.9513	0.3556	1.266	0.3
	3	0.2177	0.8115	0.1693	1.266	0.3
	4	0.1659	0.6382	0.1097	1.266	0.3

[a] $\alpha_t = 54.3$, H_2 recovered $= 0.899$, $y_{t,4} = 0.950$, $\Theta_t = 0.710$, $\Sigma St_n = 4.831$, $\eta = 0.1861$.
[b] $\alpha_t = 71.9$, H_2 recovered $= 0.925$, $y_{t,4} = 0.950$, $\Theta_t = 0.730$, $\Sigma St_n = 4.965$, $\eta = 0.1863$.
[c] $\alpha_t = 83.5$, H_2 recovered $= 0.935$, $y_{t,4} = 0.950$, $\Theta_t = 0.738$, $\Sigma St_n = 5.020$, $\eta = 0.1863$.
[d] $\alpha_t = 95.4$, H_2 recovered $= 0.944$, $y_{t,4} = 0.950$, $\Theta_t = 0.745$, $\Sigma St_n = 5.065$, $\eta = 0.1864$.

Case 2. Constant Cut: $\Theta_n = \Theta$

When the desired value of x_N, $y_{t,N}$, or product recovery is determined, assume Θ and repeate the calculation of Eq. (17) or Eqs. (26) and (27) N times. Repeat the procedure until x_N, $y_{t,N}$, or product recovery becomes close to the desired value. When the desired Θ_t is determined, calculate Θ by Eq. (13) and then calculating Eq. (17) or Eqs. (26) and (27) N times.

CALCULATION RESULTS AND DISCUSSION

The calculations were carried out using a FACOM M-380 Computer. Equations (26) and (27) were solved by Adams' method, and the iterative calculations to determine the desired performance were done by the Gold

TABLE 3
Hydrogen Separation from a H_2/CO Gas Mixture Using a Serial Fed Separator ($\alpha^* = 76.2$, $x_f = 0.75$, $N = 4$, constant cut)

Mode	n	x_n	y_n	Θ_n	St_n	Pr
Perfect mixing[a]	1	0.6657	0.9884	0.2612	0.699	0.3
	2	0.5551	0.9785	0.2612	0.722	0.3
	3	0.4166	0.9468	0.2612	1.018	0.3
	4	0.2785	0.8070	0.2612	2.334	0.3
Cocurrent[b]	1	0.6602	0.9908	0.2717	0.656	0.3
	2	0.5596	0.9834	0.2717	0.635	0.3
	3	0.3830	0.9595	0.2717	0.833	0.3
	4	0.2394	0.7680	0.2717	2.766	0.3
Cross flow[c]	1	0.6541	0.9907	0.2848	0.691	0.3
	2	0.5234	0.9825	0.2848	0.675	0.3
	3	0.3523	0.9530	0.2848	0.943	0.3
	4	0.1854	0.7712	0.2848	2.709	0.3
Countercurrent[d]	1	0.6503	0.9907	0.2930	0.713	0.3
	2	0.5129	0.9818	0.2930	0.700	0.3
	3	0.3323	0.9489	0.2930	1.014	0.3
	4	0.1496	0.7732	0.2930	2.671	0.3

[a] $\alpha_t = 49.2$, H_2 recovered = 0.889, $y_{t,4} = 0.950$, $\Theta_t = 0.702$, $\Sigma St_n = 4.773$, $\eta = 0.1863$.
[b] $\alpha_t = 60.3$, H_2 recovered = 0.911, $y_{t,4} = 0.950$, $\Theta_t = 0.719$, $\Sigma St_n = 4.889$, $\eta = 0.1863$.
[c] $\alpha_t = 83.5$, H_2 recovered = 0.935, $y_{t,4} = 0.950$, $\Theta_t = 0.738$, $\Sigma St_n = 5.020$, $\eta = 0.1863$.
[d] $\alpha_t = 108$, H_2 recovered = 0.950, $y_{t,4} = 0.950$, $\Theta_t = 0.750$, $\Sigma St_n = 5.097$, $\eta = 0.1864$.

division method. In the following calculation we used permeability data of a polyimide dense membrane which was cast on a glass plate from polymer solution and dried by heating. This polyimide was synthesized from 3,3′,4,4′-biphenyltetracarboxilic dianhydride and 4,4′-diamino-diphenyl ether (UBE Industries). Gas permeability was measured at a 0.1-MPa pressure difference using the high vacuum method, and the experimental data at 25°C are listed in Table 1.

Two examples were calculated. One is the case in which there is a concentration of H_2 up to 95% in the total permeate stream from the feed stream containing 75% H_2 and 25% CO. The other is the case in which there is a concentration of CH_4 up to 98% in the high-pressure stream from the feed stream containing 60% CH_4 and 40% CO_2.

In order to evaluate the performance of a serial fed system, we used a total separation factor α_t defined by Eq. (38) and a rating factor η defined as the recovery ratio of product per unit dimensionless membrane area, described by Eq. (39):

TABLE 4
Methane Separation from a CH_4/CO_2 Gas Mixture Using a Serial Fed Separator ($\alpha^* = 51.4$, $1 - x_f = 0.6$, $N = 4$, constant membrane area)

Mode	n	$1 - x_n$	$1 - y_n$	Θ_n	St_n	Pr
Perfect mixing[a]	1	0.8239	0.1478	0.3312	3.108	0.1
	2	0.9183	0.4193	0.1891	3.108	0.1
	3	0.9596	0.6782	0.1467	3.108	0.1
	4	0.9800	0.8346	0.1404	3.108	0.1
Cocurrent[b]	1	0.8734	0.0947	0.3512	2.253	0.1
	2	0.9373	0.4515	0.1315	2.253	0.1
	3	0.9650	0.6898	0.1006	2.253	0.1
	4	0.9800	0.8211	0.0944	2.253	0.1
Cross flow[c]	1	0.8531	0.0787	0.3268	1.804	0.1
	2	0.9347	0.3330	0.1356	1.804	0.1
	3	0.9648	0.6179	0.0869	1.804	0.1
	4	0.9800	0.7794	0.0759	1.804	0.1
Countercurrent	1	0.8362	0.0718	0.3091	1.585	0.1
	2	0.9291	0.2709	0.1411	1.585	0.1
	3	0.9636	0.5510	0.0836	1.585	0.1
	4	0.9800	0.7400	0.0685	1.585	0.1

[a] $\alpha_t = 91.42$, CH_4 recovered = 0.650, $\Theta_t = 0.602$, $\Sigma St_n = 12.43$, $\eta = 0.0523$.
[b] $\alpha_t = 127.5$, CH_4 recovered = 0.750, $\Theta_t = 0.541$, $\Sigma St_n = 9.013$, $\eta = 0.0832$.
[c] $\alpha_t = 161.1$, CH_4 recovered = 0.802, $\Theta_t = 0.509$, $\Sigma St_n = 7.217$, $\eta = 0.1111$.
[d] $\alpha_t = 184.6$, CH_4 recovered = 0.828, $\Theta_t = 0.493$, $\Sigma St_n = 6.343$, $\eta = 0.1305$.

$$\alpha_t = \frac{y_{t,N}}{1 - y_{t,N}} \frac{1 - x_N}{x_N} \tag{38}$$

$$\eta = \frac{\text{recovery ratio}}{\sum_{n=1}^{N} St_n} \tag{39}$$

Both examples were computed under the conditions that the system was constructed of four stages for constant membrane area (Case 1) and constant cut (Case 2). The results of hydrogen separation are shown in Tables 2 and 3. Total separation factors and hydrogen recovery in the countercurrent mode are better in both Case 1 (Table 2) and Case 2 (Table 3), but the rating factors are almost the same in any flow pattern. By comparing Tables 2 and 3 it is seen that the total performance of Case 1 is better than that of Case 2 in the perfect mixing and cocurrent modes, whereas the reverse is true in the countercurrent mode. There is no difference between Case 1 and Case 2 in the cross flow mode.

TABLE 5
Methane Separation from a CH_4/CO_2 Gas Mixture Using a Serial Fed Separator ($\alpha^* = 51.4$, $1 - x_f = 0.6$, $N = 4$, constant cut)

Mode	n	$1 - x_n$	$1 - y_n$	Θ_n	St_n	Pr
Perfect mixing[a]	1	0.7415	0.0790	0.2136	1.182	0.1
	2	0.8751	0.2493	0.2136	2.531	0.1
	3	0.9490	0.6032	0.2136	4.609	0.1
	4	0.9800	0.8346	0.2136	4.970	0.1
Cocurrent[b]	1	0.7289	0.0512	0.1902	0.757	0.1
	2	0.8685	0.1348	0.1902	1.333	0.1
	3	0.9485	0.5282	0.1902	3.828	0.1
	4	0.9800	0.8143	0.1902	4.718	0.1
Cross flow[c]	1	0.7074	0.0480	0.1629	0.619	0.1
	2	0.8263	0.0966	0.1629	0.889	0.1
	3	0.9312	0.2873	0.1629	1.963	0.1
	4	0.9800	0.6805	0.1629	3.748	0.1
Countercurrent[d]	1	0.6998	0.0469	0.1529	0.572	0.1
	2	0.8104	0.0874	0.1529	0.778	0.1
	3	0.9165	0.2225	0.1529	1.489	0.1
	4	0.9800	0.5644	0.1529	3.041	0.1

[a] $\alpha_t = 85.4$, CH_4 recovered = 0.625, $\Theta_t = 0.617$, $\Sigma St_n = 13.29$, $\eta = 0.0470$.
[b] $\alpha_t = 107$, CH_4 recovered = 0.702, $\Theta_t = 0.570$, $\Sigma St_n = 10.64$, $\eta = 0.0660$.
[c] $\alpha_t = 161.1$, CH_4 recovered = 0.802, $\Theta_t = 0.509$, $\Sigma St_n \approx 7.218$, $\eta = 0.1111$.
[d] $\alpha_t = 200.3$, CH_4 recovered = 0.841, $\Theta_t = 0.485$, $\Sigma St_n \approx 5.879$, $\eta = 0.1431$.

The results of methane separation are shown in Tables 4 and 5. The differences in total performance between the four flow patterns are more obvious. By analysis of the single-stage operation, the limiting value of methane concentration is 95% in the perfect mixing mode and 96% in the cocurrent flow mode. However, by using the serial fed system consisting of more than two modules, methane can be concentrated up to 98% in these flow patterns.

Analysis on Effect of Flow Pattern and Stage Number

The effect of flow pattern and stage number on the performance of the serial fed multistage system were examined for the above two examples. The calculated results are illustrated in Fig. 4 for hydrogen separation and in Figs. 5 and 6 for methane separation.

It is known that separation efficiency of the single module is best in the

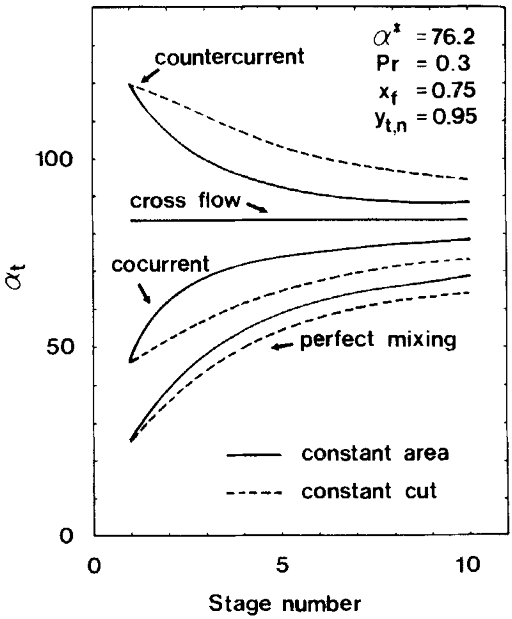

FIG. 4. Effect of flow pattern and stage number on the total separation factor in the case in which there is concentrate H_2 up to 95% in the total permeate stream from the 75% H_2 and 25% CO mixture.

countercurrent mode and becomes lower in the following order: cross flow, cocurrent, perfect mixing. Figures 4 and 5 show that total separation factors depend on the effect of flow pattern in a single module and become closer to that of cross flow with an increase of the stage number. It is recognized intuitively that an infinite number of stages in this system is essentially identical with the cross flow model. Consequently, a system consisting of countercurrent flow modules is disadvantageous and so it is preferable to construct a system with the smallest number of stages possible by using large modules. If difficulties in operation are to be eliminated, it is desirable to assemble a parallel fed multiple stage system containing countercurrent flow modules. On the other hand, it is advantageous to construct a serial fed system made up of cocurrent flow or perfect mixing modules, and to increase the stage number by using small modules.

Rating factors of the hydrogen separation example were approximately 0.1863 regardless of the flow pattern and the number of stages. However,

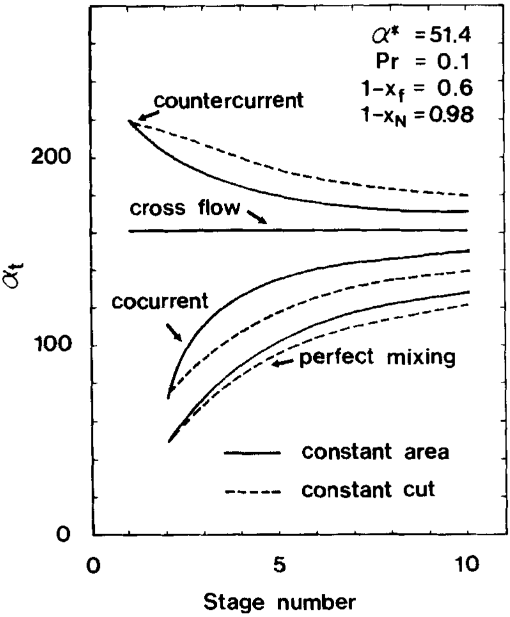

FIG. 5. Effect of flow pattern and stage number on the total separation factor in the case in which there is concentrate CH_4 up to 98% in the high-pressure stream from the 60% CH_4 and 40% CO_2 mixture.

in the methane separation example the rating factors were strongly dependent on the flow pattern and the number of stages as illustrated in Fig. 6. This means that application of a serial fed system to the case in which the product is concentrated in the high-pressure stream is more strongly influenced by the number of stages and the flow pattern.

Figures 4, 5, and 6 give other information: It is better to construct a system composed of countercurrent flow modules in the constant cut mode than in the constant membrane area mode. On the other hand, it is better to use a system consisting of perfect mixing or cocurrent flow modules in the constant membrane area mode.

From a practical standpoint, flow in a membrane module is not always ideal flow, and its performance lies between those of perfect mixing and countercurrent flow. However, the calculation methods proposed in this study are able to predict the performance of a serial fed system by selecting the flow pattern closest to the actual one from among the four kinds of ideal flow modes.

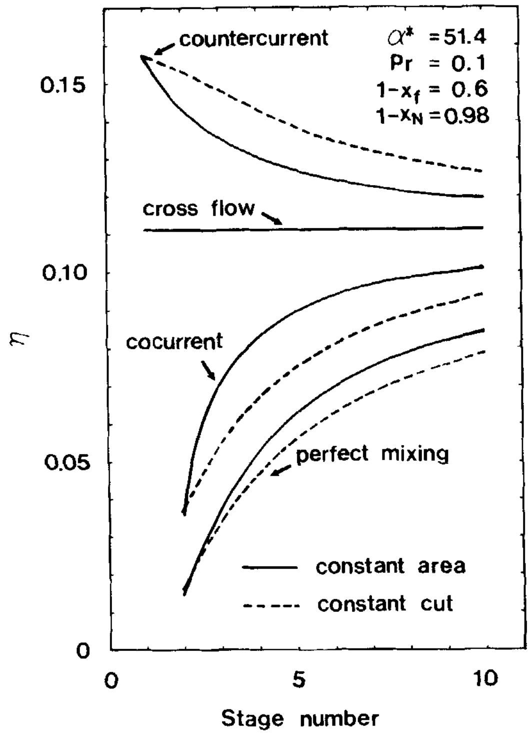

FIG. 6. Effect of flow pattern and stage number on the rating factor in the case in which there is concentrate CH_4 up to 98% in the high-pressure stream from the 60% CH_4 and 40% CO_2 mixture.

CONCLUSION

1) One can easily design a serial fed multistage system for the separation of a binary gas by using the calculation methods presented.

2) The system consisting of countercurrent flow modules for constant cut is the most efficient among the cases studied, and should be designed with the smallest number of stages possible.

3) For a perfect mixing or cocurrent flow module, it is favorable to design the system in the constant membrane area mode and to increase the number of stages by using small modules.

4) The performance of a system composed of cross flow modules is identical with that of a single module.

SYMBOLS

F	flow rate in high pressure stream ($m^3 \cdot s^{-1}$)
F'	dimensionless flow rate defined by Eq. (28)
L	flow rate in permeate stream ($m^3 \cdot s^{-1}$)
l	thickness of membrane (m)
N	total stage number
Pr	pressure ratio of low to high (p_l/p_h)
p	total pressure (Pa)
Q	gas permeability ($m^3(STP) \cdot m \cdot m^{-2} \cdot s^{-1} \cdot Pa^{-1}$)
S	actual membrane area (m^2)
St	dimensionless membrane area
S^*	capacity factor defined by Eq. (21) (m^2)
T	flow rate in total permeate stream ($m^3 \cdot s^{-1}$)
x	mole fraction of faster permeating gas in high-pressure stream
y	mole fraction of faster permeating gas in permeate stream

Greek

α_t	total separation factor defined by Eq. (38)
α^*	permeability ratio (Q_A/Q_B)
η	rating factor defined by Eq. (39)
Θ	stage cut
Θ_t	total cut defined by Eq. (9)

Subscripts

A	refers to faster permeating gas
B	refers to later permeating gas
f	refers to inlet of first stage concerning composition
h	refers to high-pressure side
l	refers to low-pressure side
n	refers to nth stage
t	refers to total
0	refers to inlet of first stage concerning flow rate

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Received by editor November 6, 1984.